

# Spin relaxation near the metal-insulator transition: dominance of the Dresselhaus spin-orbit coupling

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We identify the Dresselhaus spin-orbit coupling as the source of the dominant spin-relaxation mechanism in the impurity band of doped semiconductors. The Dresselhaus-type (i.e. allowed by bulk-inversion asymmetry) hopping terms are derived and incorporated into a tight-binding model of impurity sites, and they are shown to unexpectedly dominate the spin relaxation, leading to spin-relaxation times in good agreement with experimental values. This conclusion is drawn from two complementary approaches employed to extract the spin-relaxation time from the effective Hamiltonian: an analytical diffusive-evolution calculation and a numerical finite-size scaling.

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Spin dynamics in semiconductors is a fundamental issue in view of the rich physics involved and the potential technological applications [1, 2]. It is thus not surprising that spin relaxation studies were already performed in the early days of semiconductor research [3–5] and are intensely pursued today with modern experimental techniques [6, 7]. An intriguing experimental observation is the fact that in n-doped semiconductors at low temperatures the spin relaxation time  $\tau_s$  presents a maximum as a function of the doping density near the metal-insulator transition (MIT) [5, 6, 8–12].

Interestingly, while the mechanisms behind spin relaxation have been properly identified at high temperatures or for doping densities away from the critical one [10, 13, 14], a theoretical understanding of low-temperature spin relaxation close to the MIT is still lacking. This unsatisfactory state of affairs has motivated some attempts to identify the relevant mechanisms for spin relaxation [15–18] close to the MIT. In particular, on the metallic side of the transition, Shklovskii has proposed the applicability of the well-known Dyakonov-Perel mechanism usually valid in the conduction band [15]. Furthermore, a tight-binding model of impurities including Rashba spin-orbit coupling has been developed [18]. The spin relaxation times resulting from this last model were larger than the experimental values, implying that other mechanisms should be active in this density regime.

In this work we identify the Dresselhaus coupling as the source of the leading spin relaxation mechanism on the metallic side of the transition. This conclusion is based on the construction of an effective spin-orbit Hamiltonian for the impurity system of III-V semiconductors, together with its analytical and numerical solution. The resulting spin relaxation times are in good agreement with the existing experimental values for GaAs. The detailed temperature-dependent measurements of Ref. [12] yielded a saturation of  $\tau_s$  below 10 K, indicating that inelastic processes are irrelevant at low temperatures. We thus work with a zero-temperature formalism.

The envelope-function approximation (EFA) for describing conduction-band electrons in zincblende semiconductors in-

corporates the lattice-scale physics (described by the periodic part of the Bloch wave function) into the effective one-body Hamiltonian [19, 20]

$$H = H_0 + H_{\text{SIA}} + H_{\text{BIA}} \quad (1)$$

$$H_0 = \frac{p^2}{2m^*} + V(\mathbf{r}) \quad (2)$$

$$H_{\text{SIA}} = \lambda \boldsymbol{\sigma} \cdot \nabla V \times \mathbf{k} \quad (3)$$

$$H_{\text{BIA}} = \gamma [\sigma_x k_x (k_y^2 - k_z^2) + \text{cyclic permutations}]. \quad (4)$$

The electrostatic potential  $V(\mathbf{r})$  includes all potentials aside from the crystal one, while  $\boldsymbol{\sigma}$  is the vector of Pauli matrices and  $\mathbf{k} = \mathbf{p}/\hbar$ . The effective spin-orbit coupling  $\lambda$ , enabled by the structural inversion asymmetry (SIA) is usually orders of magnitude larger than the one of vacuum  $\lambda_0 = \hbar^2/4m_0^2c^2 \simeq 3.7 \times 10^{-6} \text{ \AA}^2$ . It can be calculated at the level of the 8-band Kane model, which, for example, for GaAs yields  $\lambda \simeq -5.3 \text{ \AA}^2$  [20]. The bulk inversion asymmetry (BIA) coupling constant  $\gamma$  is another material-dependent parameter. The exact value of  $\gamma$  is a matter of current debate [1, 20]. A 14-band model is required for the theoretical estimation of  $\gamma$ , leading to  $\gamma \approx 27 \text{ eV \AA}^3$  for GaAs [20, 21]. More refined theoretical calculations yield somewhat lower values [22–24]. While early experimental values obtained in bulk samples agree approximately with the above-quoted value of  $27 \text{ eV \AA}^3$  [25], recent results inferred from measurements in low-dimensional systems are again consistently lower [1, 26–28].

In order to study the spin relaxation in the impurity band near the MIT, we consider the potential  $V(\mathbf{r})$  due to the ionized impurities, given by

$$V(\mathbf{r}) = \sum_p V_p(\mathbf{r}) = - \sum_p \frac{e^2}{\epsilon |\mathbf{r} - \mathbf{R}_p|}, \quad (5)$$

where  $\epsilon$  is the dielectric constant of the semiconductor and  $\mathbf{R}_p$  represents the impurity positions. The potential  $V_p$  gives rise to hydrogenic states centered at the impurity  $p$ . In order to build the basis of electronic states we will only con-

sider the ground state  $\phi_p(\mathbf{r}) = \phi(|\mathbf{r} - \mathbf{R}_p|)$ , with  $\phi(\mathbf{r}) = (1/\sqrt{\pi a^3}) \exp(-r/a)$ , and  $a$  the effective Bohr radius.

The second-quantized form of the Hamiltonian (1), that we denote  $\mathcal{H}$ , has components

$$\mathcal{H}_0 = \sum_{m \neq m', \sigma} \langle m' \sigma | H_0 | m \sigma \rangle c_{m' \sigma}^\dagger c_{m \sigma}, \quad (6)$$

$$\mathcal{H}_{\text{SO}} = \sum_{m \neq m', \sigma} \langle m' \bar{\sigma} | H_{\text{SO}} | m \sigma \rangle c_{m' \bar{\sigma}}^\dagger c_{m \sigma}, \quad (7)$$

where the label SO stands for SIA or BIA. We denote the 1s state  $\phi_m(\mathbf{r})$  with spin  $\sigma = \pm 1$  in the z-direction by  $|m \sigma\rangle$ , and  $c_{m \sigma}^\dagger$  ( $c_{m \sigma}$ ) is the creation (annihilation) operator of a particle in that state ( $\bar{\sigma} = -\sigma$ ). The matrix elements in Eq. (6) contain three-center integrals  $\langle m' \sigma | V_p | m \sigma \rangle$  with  $p \neq m$ . Due to the exponential decay of  $\phi_m(\mathbf{r})$ , one usually keeps only the term

$$\langle m' \sigma | V_{m'} | m \sigma \rangle = -V_0 \left(1 + \frac{R_{m'm}}{a}\right) e^{-R_{m'm}/a}, \quad (8)$$

with  $V_0 = e^2/\varepsilon a$  (twice the binding energy of an isolated impurity) and  $R_{m'm}$  the distance between the two impurities. The resulting Hamiltonian  $\mathcal{H}_0$  defines the well-known Matsubara-Toyozawa model (MT) [29], which has been thoroughly studied in the context of the MIT. The subtleties, drawbacks and applicability of this model to describe the metallic side of the MIT, as well as its extension to include the  $\mathcal{H}_{\text{SIA}}$  spin-orbit coupling, have recently been discussed [30]. Electron-electron interactions induce significant many-body effects on the insulating side of the transition, but not on the metallic side. Therefore we do not need to include them in our model. According to the Mott criterion the critical dimensionless impurity density for the MIT corresponds to  $\mathcal{N}_i = n_i a^3 \simeq 0.017$ , corresponding to a critical density of  $2 \times 10^{16} \text{ cm}^{-3}$  for GaAs.

The matrix element of  $\mathcal{H}_{\text{SIA}}$  is

$$\langle m' \bar{\sigma} | H_{\text{SIA}} | m \sigma \rangle = \frac{\sigma \lambda}{a^2} \int d\mathbf{r} V(\mathbf{r}) \frac{\phi_{m'}(\mathbf{r}) \phi_m(\mathbf{r})}{|\mathbf{r} - \mathbf{R}_{m'}| |\mathbf{r} - \mathbf{R}_m|} \times [(z - z_m)(r_\sigma - R_{m'\sigma}) - (z - z_{m'})(r_\sigma - R_{m\sigma})], \quad (9)$$

where  $r_\sigma = x + i\sigma y$  and  $R_{m\sigma} = X_m + i\sigma Y_m$ . The Hamiltonian  $\mathcal{H}_{\text{SIA}}$  represents the generalization of the Rashba coupling to the case of impurity potentials, and was introduced in Ref. [18]. There, an alternative path to the EFA was followed in order to calculate the matrix elements  $\langle m' \bar{\sigma} | H_{\text{SIA}} | m \sigma \rangle$ , which made use of impurity states with spin admixture obtained from spin-admixed conduction-band Bloch states derived at the level of the 8-band Kane model. We remark that the terms corresponding to  $p = m, m'$  in  $V(\mathbf{r})$  give vanishing contributions to the SIA matrix element due to the axial symmetry of the two-center integrals. As a consequence, the Rashba matrix elements are given by three-center integrals, resulting in very slow spin relaxation [18] in comparison with experimental results. We therefore turn to the Dresselhaus term, whose matrix element is given by

$$\begin{aligned} \langle m' \bar{\sigma} | H_{\text{BIA}} | m \sigma \rangle &= \gamma [\langle m' | k_x (k_y^2 - k_z^2) | m \rangle \\ &\quad + i \sigma \langle m' | k_y (k_z^2 - k_x^2) | m \rangle] \\ &= \frac{\gamma}{\pi a^3} (\sigma I_{y,m'm} + i I_{x,m'm}), \end{aligned} \quad (10)$$

where

$$I_{x,m'm} = \frac{1}{a^3} \int d\mathbf{r} \frac{e^{-|\mathbf{r} - \mathbf{R}_{m'm}|/a} e^{-r/a}}{|\mathbf{r} - \mathbf{R}_{m'm}| r^3} \times (a + r)(x - X_{m'm})(y^2 - z^2), \quad (11)$$

and  $I_{y,m'm}$  is obtained from  $I_{x,m'm}$  with the exchanges  $X_{m'm} \leftrightarrow Y_{m'm}$  and  $x \leftrightarrow y$ . Performing a rotation of the coordinate system and switching to cylindrical coordinates allows us to easily do the angular integral, yielding

$$I_{x,m'm} = \frac{\pi c}{a^3} \int d\rho dz \rho \frac{e^{-\sqrt{\rho^2 + (z - R_0)^2}/a} e^{-\sqrt{\rho^2 + z^2}/a}}{\sqrt{\rho^2 + (z - R_0)^2} (\rho^2 + z^2)^{3/2}} \times \left(a + \sqrt{\rho^2 + z^2}\right) \left[\frac{\rho^2}{2} (3z - R_0) - z^2 (z - R_0)\right], \quad (12)$$

where

$$c = 2 \cos \varphi \sin \theta [1 - \sin^2 \theta (1 + \sin^2 \varphi)], \quad (13)$$

and  $(R_0, \theta, \varphi)$  are the polar coordinates of  $\mathbf{R}_{m'm}$  in the original reference frame. The integrals in (12) are not analytically solvable, but they can easily be integrated numerically. This is the route that we take later, where we simulate the dynamical evolution of initially prepared pure spin states. Before tackling the numerical problem, we provide a simple estimation of the spin lifetime based on the steepest-descent approximation of the integral (12) (valid in the limit  $R_0 \gg a$ ), given by

$$I_{x,m'm} \simeq \frac{\pi^2 c}{4a^2} R_0 \left(a + \frac{R_0}{2}\right) e^{-R_0/a}. \quad (14)$$

The spatial diffusion of electrons through the network of impurities is accompanied by a spin diffusion characterized by a typical spin rotation angle per hop [18]

$$\langle \Theta^2 \rangle = \frac{15}{2} \frac{\sum_{m \neq m'} |\langle m' \bar{\sigma} | H_{\text{BIA}} | m \sigma \rangle|^2}{\sum_{m \neq m'} |\langle m' \sigma | H_0 | m \sigma \rangle|^2}. \quad (15)$$

The spin-relaxation time can be defined as the time in which the accumulated rotation reaches a unit angle, and therefore is given by

$$\frac{1}{\tau_s} = \frac{2}{3} \frac{\langle \Theta^2 \rangle}{\tau_c}, \quad (16)$$

where  $\tau_c$  is the typical hopping time. It can be shown that this expression is independent of whether the initial state is localized or extended [18]. Taking  $\tau_c$  as the time needed for the initial-state population on an impurity site to drop from 1 to 1/2, we obtain

$$\frac{1}{\tau_c} = \frac{\sqrt{2}}{\hbar} \left( \sum_{m \neq m'} |\langle m' \sigma | H_0 | m \sigma \rangle|^2 \right)^{1/2} \simeq \frac{\sqrt{14\pi} V_0}{\hbar} \mathcal{N}_i^{1/2}. \quad (17)$$

For the second equality [32] we have used the impurity average assuming a random distribution without hard-core repulsive effects on the scale of the effective Bohr radius [18, 31]. Performing also the impurity average in Eq. (15) we have

$$\langle \Theta^2 \rangle = 0.258 \left( \frac{\gamma}{a^3 V_0} \right)^2, \quad (18)$$

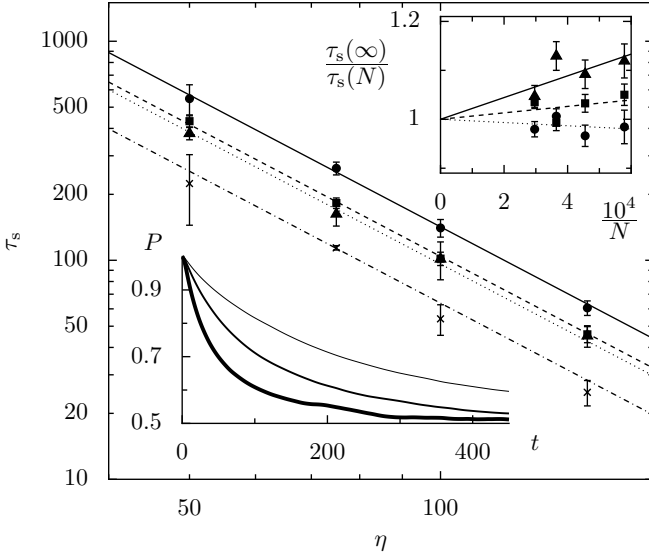


FIG. 1. Scaling of the spin relaxation time extrapolated to infinite system size, with the spin-orbit enhancement factor  $\eta$ , for densities  $N_i = 0.02$  (circles),  $0.029$  (squares),  $0.037$  (triangles), and  $0.06$  (crosses). The lines are fits of a quadratic dependence of the relaxation rate on  $\eta$ . The times are given in units of  $\hbar/V_0$ . *Lower Inset*: Spin survival probability  $P$  for an initial MT eigenstate evolving under the enhanced Dresselhaus couplings for a density  $N_i = 0.029$  and a system size  $N = 3375$ . Lines of increasing thickness are for enhancement factors  $\eta = 75, 100$ , and  $150$ . *Upper Inset*: Size dependence of  $\tau_s^{-1}$  for  $\eta = 150$ . Lines are linear fits to the data that allow to extrapolate to the infinite-size values.

and

$$\frac{1}{\tau_s} = \frac{1.14 \gamma^2}{a^6 V_0 \hbar} N_i^{1/2}. \quad (19)$$

This expression for the spin relaxation time is our main result. As we discuss below, its confrontation with the experimental data allows us to identify the Dresselhaus coupling as the dominant spin relaxation mechanism in the regime of study. Furthermore, numerical calculations of the relaxation time within our model provide a complementary path validating the analytical approach, since some of the previously used approximations can be avoided.

The numerical procedure starts from the numerical integration of (12) for a given impurity configuration in order to obtain the matrix elements (10) for  $H_{\text{BIA}}$  (and similarly for  $H_{\text{SIA}}$ ). We then diagonalize the total Hamiltonian  $\mathcal{H}$  including the two contributions to the spin-orbit coupling, which allows us to obtain the quantum evolution of an arbitrary state. Choosing an initial state with a well-defined spin projection (for instance a MT eigenstate with  $\sigma = 1$ ) we can follow the spin evolution and extract the spin lifetime from it. The weakness of the spin-orbit coupling translates into spin-admixture perturbation energies which are, even for the largest system sizes (in terms of number of impurities,  $N$ ) that we are able to treat numerically, orders of magnitude smaller than the typical MT level spacing. This large difference between the two

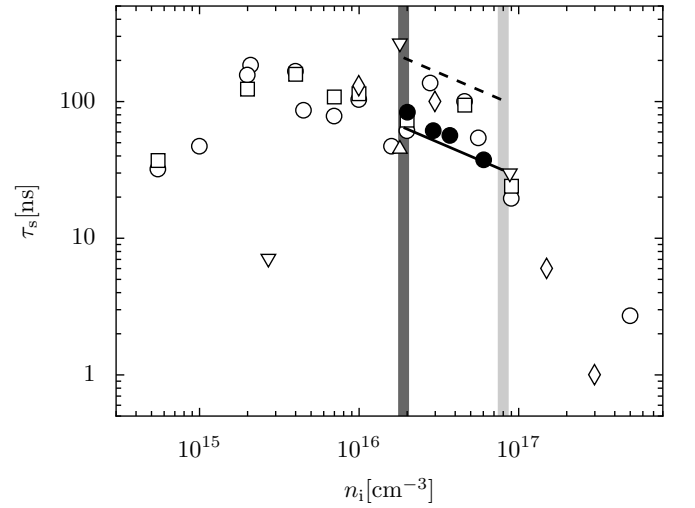


FIG. 2. Spin relaxation time in n-doped GaAs as a function of the doping density. The prediction of Eq. (19) (solid line) and our numerical results (filled circles) for the regime between the metal-insulator transition (dark gray line) and the hybridization of the impurity band with the conduction band (light gray line) obtained using  $\gamma = 27 \text{ eV \AA}^3$  are compared to experimentally obtained values (open symbols) using different methods. Circles and squares are from Ref. [10] for  $T = 2 \text{ K}$  and  $T = 4.2 \text{ K}$ , respectively, along with data from Ref. [6] (diamonds), [7] (triangles), and [12] (reversed triangles). The dashed line represents the result of Eq. (19) with  $\gamma = 15 \text{ eV \AA}^3$ .

energy scales in finite size simulations masks the spin-orbit-driven physics, and forces us to follow an indirect path: we introduce an artificially enhanced coupling constant  $\eta\gamma$  and a finite-size scaling procedure. The limits  $N \rightarrow \infty$  and then  $\eta \rightarrow 1$  taken at the end of the calculation provide the sought spin-relaxation rate. The numerically extracted values of the spin relaxation times associated with  $H_{\text{SIA}}$  are, consistently with the results of Ref. [18], considerably larger than the ones experimentally observed. Therefore, we hereafter neglect the Rashba term in the numerical calculations, and concentrate on the spin evolution governed by  $H_{\text{BIA}}$ .

In the lower inset of Fig. 1 we show typical spin evolutions starting from an eigenstate of the MT system with  $\sigma = 1$  in the energy range of extended states of the impurity band, for a density  $N_i = 0.029$  just above the MIT transition for three values of the coupling constant and  $N = 3375$  impurities. The initial perturbative regime with a quadratic time decay of the spin survival is followed by an exponential decay from which the relaxation rate  $\tau_s^{-1}$  can be inferred, until the saturation value of  $1/2$ . For each density and effective coupling constant  $\eta\gamma$  the asymptotic value of  $\tau_s^{-1}$  can be obtained by extrapolating the finite- $N$  values to the infinite size limit (upper inset of Fig. 1). We ran a sufficiently large number of impurity configurations (typically 40) to make the statistical error negligible [33]. The resulting error bars arise from the quality of the fittings to the exponential decay of the spin survival. In agreement with our analytical results, an inverse quadratic dependence of  $\tau_s$  on the coupling strength is obtained (Fig.

1). Fitting this dependence of  $\tau_s$  on  $\eta$  allows us to extract the physical values ( $\eta = 1$ ) of  $\tau_s$ .

In Fig. 2 we present the spin relaxation times resulting from our numerical approach for GaAs at four different impurity densities above the MIT (black dots), together with the prediction of Eq. (19) (solid line), and the available experimental data from Refs. [6, 7, 10, 12]. The agreement between the analytical and numerical approaches is very important in view of the complementarity of these two very different ways to extract  $\tau_s$ . Both approaches describe the data within the experimental uncertainty and correctly reproduce the density dependence of the spin relaxation time. The small departure of the analytical and numerical results is not significant, taking into account the different approximations of both paths and the arbitrariness associated with the definition of relaxation times, i.e. numerical prefactors in Eqs. (16) and (17).

While in the critical region and deep into the localized regime there is some dispersion of the experimental values for GaAs, depending on the different samples and measurement technique, on the metallic side of the MIT values of  $\tau_s \geq 100$  ns are consistently obtained. A decrease of  $\tau_s$  with  $n_i^{1/2}$  is observed, with a clear change in the density-dependence once the impurity and the conduction bands hybridize. Our analytical and numerical results of Fig. 2 (solid line and filled symbols) are obtained using the values  $V_0 = 11.76$  meV and  $\gamma = 27$  eVÅ<sup>3</sup> without any adjustable parameter. We remark that these results are very sensitive to the value of  $\gamma$ . For instance, taking  $\gamma = 15$  eVÅ<sup>3</sup>, that is proposed by some measurements [1, 26], results in the dashed line. The identification of the Dresselhaus coupling as the dominant channel for spin relaxation close to the MIT of 3-dimensional samples will help in the precise determination of the material constant  $\gamma$ , which is crucial for the operation of low-dimensional spintronic devices.

In conclusion, we have identified a spin relaxation mechanism characteristic of electrons on the metallic side of the metal-insulator transition in the impurity band of semiconductors, where up to now a theoretical understanding was lacking, thereby solving a longstanding problem in spintronics. Our mechanism is derived from the Dresselhaus spin-orbit coupling. It dominates over the usually stronger Rashba coupling in the landscape of hydrogenic impurities in semiconductors with zincblende structure, and provides relaxation times that agree with the experimentally measured values.

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- [32] We have obtained an excellent agreement between Eq. (17) and the numerically computed time-evolution of an initially localized state within the MT model.
- [33] The  $\eta$ - and  $N$ -dependent values of  $\tau_s$  do not depend on whether we extract them from the impurity-averaged spin survival curve or by averaging the rates of different configurations.